### Title
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**Authors response to the above comment by M. Vogt et al. on "New Directions: Enhancing the natural cycle to slow global warming"** ☆

We appreciate the interest by our colleagues (Vogt et al., 2008) in Wingenter et al. (2007), in which we presented a possible means of lowering the temperature over the Southern Ocean (SO) region. The best application of our proposal may be to cool the surface temperature, which would reduce sea-level rise (i) by slowing sea water expansion; and (ii) by diminishing the breakup of Antarctic coastal ice shelves, which buttress glacial flow into the SO. Our paper had two main intentions: first to encourage modeling studies in this direction, and secondly to show that large-scale iron fertilization for the purpose of carbon sequestration could have severe negative impacts by overcooling the region. Vogt et al. raised several concerns about our initial calculations, which we clarify below.

Our proposed iron fertilization of 2% of the SO was estimated to "result in a 2 °C decrease in temperature over the SO region". Vogt et al. have extrapolated similar results globally and annually, to suggest an impact on the order of 0.005 °C on these larger scales. While we do not dispute the general validity of this scaled up calculation, the assertion that the cooling effect of our proposed estimate is therefore a factor of 400 too large (i.e. $2 \div 0.005$) is incorrect, because we never stated that the calculated 2 °C decrease applied to the entire planet for 12 months. Instead, the 2 °C decrease that we estimated over the SO region is a factor of three higher than the 0.6 °C determined by Vogt et al. for this region.

The factor of three difference that remains between the two estimates mostly results from disagreement over the amount of DMS which will increase over the SO (20% in Wingenter et al. vs. 10% in Vogt et al., or a factor of two). Of the three published SO iron experiments that made DMS measurements, the response that we used (a 480% increase in DMS after fertilization)—from our SOFeX North observations (Wingenter et al., 2004)—was in the middle of the three experiments (Table 1). The three published responses range from 170–680%, and we agree that the uncertainty in the ocean response ($480 \pm 250\%$; a 50% uncertainty) cannot be neglected. Considering the relatively limited surface area covered by each of these experiments, the much larger area in our proposal would probably exhibit heterogeneous responses, i.e. higher than the SOFeX North response is some areas, and lower in others. It should also be noted that the three published SO iron fertilization experiments listed in Table 1 indicate that the initial background chlorophyll *a* and DMS concentrations were low. In the proposed experiment, iron would not be added indiscriminately to areas already in bloom, as Vogt et al. suggest. Instead, satellite observations of chlorophyll *a* in conjunction with fast *in situ* biological indicators such as chlorophyll fluorescence and DMS would be used to direct the limited fertilization and maximize the impact of the iron distributed.

Table 1
Initial, outside-the-patch, and maximum-inside-the-patch DMS concentrations

| Experiment | [DMS] (nM) | | | | [Chlor *a*] (mg m$^{-3}$) | |
|---|---|---|---|---|---|---|
| | Initial DMS | Out | Max in | Change (%) | Initial or out | Inside |
| SOIREE[a] | 0.5 | 0.5 | 3.4 | 680 | 0.25 | 2.3 |
| EISENEX[a] | 1.9 | 1.8 | 3.1 | 170 | 0.5 | 2.8 |
| SOFeX N[b] | | 1.6 | 7.7 | 480 | 0.2 | 2.4 |
| SOFeX S[c,d] | | | | Increased[c,d] | 0.2 | 3.7[e] |

[DMS] is in nM, and the % changes are from the published iron addition experiments in the SO region.
DMS references.
[a]Turner et al., 2004.
[b]Wingenter et al., 2004.
[c]Wingenter, unpublished data.
[d]DMS was observed for only 11 days after the initial iron application, after which the Revelle headed back to the North patch. The peak in DMS was never observed.
[e]The maximum in chlorophyll *a* was observed 46 days after the initial fertilization on the Melville (Coale et al., 2007, Science 304, 408–414).

An additional area of disagreement relates to the calculations that determine the extent of cooling following DMS enhancements. Keys are (i) the extent to which *total* condensation nuclei (CCN) increase following a 20% increase in DMS (10% in Wingenter et al. vs. 2.6% in Vogt et al.), and (ii) the number density of total CCN that is used in the calculations (400 CCN $cm^{-3}$ in Wingenter et al. vs. 100 CCN $cm^{-3}$ in Vogt et al.). The relative increase in additional CCN enhances cloud albedo by either 3 W $m^{-2}$ (Wingenter et al.) and 0.8 W $m^{-2}$ (Vogt et al.), to give estimated temperature decreases of 2 and 0.6 °C, respectively.

We do not agree that fertilizing 2% of the SO will result in only to a 2.6% increase in total CCN. Our original estimate predicts a 20% increase in DMS would lead to a 10% increase in CCN when applying an analysis similar to that of Watson and Liss (1998). Wingenter et al. assessed the impact of additional DMS flux on albedo by following the example of Watson and Liss and assuming that initially 200 CCN $cm^{-3}$ came from sea salt and another 200 $cm^{-3}$ originated from non-sea salt sulfate aerosols derived from DMS. A 20% increase in DMS should result in a 20% more CCN derived from DMS oxidation (Wingenter et al.), or a 10% increase in the total CCN (from 400 to 440 CCN $cm^{-3}$). Vogt et al. suggest another way to treat the impact of additional DMS on CCN concentration using the linear relationship found in Ayers et al. (1997), where the monthly mean DMS mixing ratios and monthly mean CCN levels are described by $[CCN]_{normalized} = 0.52[DMS]_{normalized} + 0.48$. It is important to realize that by this correlation, a 20% increase in DMS leads to a 10% increase in *total* CCN, which is equivalent to a 20% increase in DMS-derived CCN if half of the CCN were derived from DMS. Fortuitously, this is the same increase in CCN derived in Wingenter et al., using the treatment of Watson and Liss. However, Vogt et al. using the same equation came up with only 2.6% increase in CCN for a 2% fertilization of the SO. Half of this can be explained by the factor-of-two difference in DMS response to iron fertilization; but it appears they applied both Ayers' equation and Watson and Liss' treatment. However, Ayers et al. treatment includes all CCN, not just non-sea salt sulfate particles.

Using the equation in Ayers et al., instead of a treatment similar to that in Watson and Liss, makes the initial number and type of CCN irrelevant because the cloud susceptibility or thickness is adjusted to the observed cloud albedo (Twomey, 1991). However, we continue to examine the concentration of CCN over the SO because the relationship between sulfate particles, sea-salt CCN and new particle growth may be important in future studies. During the First Aerosol Characterization Experiment (ACE-1), which used land, ship and aircraft measurements near Tasmania and over the SO from 40°S to 55°S, new particle formation was not observed in the marine boundary layer. Rather, newly nucleated sulfate particles were observed as a result of cloud processing in regions with low aerosol surface area, with the source of the sulfate being DMS (Bates et al., 1998; Clarke et al., 1998). During the experiment, Bates et al. observed about 300 total CCN $cm^{-3}$on average under background conditions, of which about 270 CCN particles were from non-sea salt sulfate and 30 particles were from sea salt. Hudson et al. (1998) also measured total CCN (saturation of 1.2%) of about 220 CCN $cm^{-3}$ during ACE-1 south of Tasmania under clean marine conditions. At Cape Grim the average total CCN concentration under clean marine conditions in the summer is about 300 CCN $cm^{-3}$ (Ayers et al., 1997). However, we note that most of these CCN are non-sea salt sulfate particles, and none of these estimates include ultrafine or new nucleated particles, which are also derived from DMS. If we assume 300 CCN $cm^{-3}$ initially, a 20% increase in DMS flux, and we apply Ayers' equation, then the final CCN number would increase from 300 to 330 CCN $cm^{-3}$. This would result in an albedo increase from 46%, using the treatment, to 46.8%, or an extra 3 $Wm^{-2}$ reflected off to space—the same result as determined in Wingenter et al.

It should be noted that there are several uncertainties in new particle production from molecular precursors, cloud formation, albedo and cloud microphysics. These are areas of active research that have been ongoing for many years. As noted in Wingenter et al., a large-scale experiment should greatly add to our understanding of aerosol growth and formation in the SO region. Careful, controlled application of our proposal may be an important test of the CLAW hypothesis.

Other major questions related to this proposal are how long would DMS production continue and how much additional DMS would be released into the atmosphere? Does the additional DMS come as a pulse over a few days after iron fertilization or would the additional DMS flux be spread out over a few weeks? For example, chlorophyll *a* persisted

during SOIREE for at least 40 days and both SOFeX North and South iron-enriched patches also persisted about 40 days but would DMS production last this long?

It is difficult to estimate the individual uncertainties of each process in our proposal. If each step in our analysis introduces a factor of two uncertainty, then the propagated error would be about a factor of four. Thus, even though the difference in temperature cooling between our estimate (2 °C) vs. that of Vogt et al.'s (0.6 °C) Vogt et al.'s is quantifiable, the factor of three difference is within the estimated uncertainty. After full-scale modeling and initial field experiments are performed, the amount of area fertilized can be adjusted to achieve the desired result. The estimate by Vogt et al. suggests that we would need to fertilize 6% of the SO to achieve a 2 °C temperature decrease over the region. However, we suggest a more conservative approach, starting at the low end of the range. Furthermore, a 2 °C decrease in temperature may be too severe. This amount was proposed merely for illustrative purposes, a starting point for discussion, whereas a 0.5 °C cooling may accomplish the intended effect with fewer unintended perturbations.

Once these questions are better understood and our goals are more clearly recognized, we will know how frequently to fertilize with iron and in what areas. Stabilizing Antarctic ice shelves may be the best application of limited iron fertilization and may require less iron over a shorter time each season and perhaps in a smaller area closer to Antarctica. Use of fully coupled models would aid in the planning and would precede such a task. We reiterate that full-scale iron fertilization for the purpose of carbon sequestration will (i) overcool the region and (ii) would introduce about 20 times more iron to the region. Any plan to fertilize the SO must first address the issue of overcooling. Because the scales and goals of the full and limited iron fertilization proposals are very different, each proposal must be weighed on its own merits and weaknesses. At this point we are only advocating modeling studies, which we hope scientific organizations will endorse. Climate change is a serious problem. The enduring solution will ultimately come when greenhouse gas emissions are controlled. However, to date $CO_2$ concentration increases are accelerating. Therefore, we need to explore and evaluate possible Climate Engineering solutions, which could counter, for the short term (a few decades), some effects of global warming such as sea level rise.

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